

PATENT Customer No. 22,852

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Protest of Application of:)
William DOWNS et al.) Group Art Unit: Unknown
Application No.: 10/591,855 (U.S. National Stage of International Application No. PCT/US2005/009590)	Examiner: Unknown
International Filing Date: March 21, 2005	GREGORY MILLS
U.S. National Stage Filing Date: September 5, 2006) QUALITY ASSURANCE SPECIALIST
For: BROMINE ADDITION FOR THE IMPROVED REMOVAL OF MERCURY FROM FLUE GAS	DEC 2 8 2006
	['] TC 1700

Commissioner for Patents P.O. Box 1450 Alexandria, VA 22313-1450

PROTEST UNDER 37 C.F.R. § 1.291(a)

Pursuant to 37 C.F.R. § 1.291(a), this paper is submitted in protest of the issuance of any

U.S. patent which claims priority to or is based on International Application No.

PCT/US2005/009590 ("the '590 application"), which was published by the International Bureau on October 6, 2005, as WO 2005/092477 ("the '477 publication"). See Exhibit A. It is believed that at least one application for patent claiming priority to or based on the international '590 application is pending in the U.S. at the time this Protest is being filed (hereinafter "the pending protested application") because, based on information provided by the U.S. Patent Office to the undersigned, a national stage application was received by the U.S. Patent Office on September 5, 2006, which is based on or claims priority to the international '590 application, and which is believed to have been accorded U.S. Application No. 10/591,855.

I. BASIS OF PROTEST IN ACCORDANCE WITH 37 C.F.R. § 1.291(B)

A. <u>Timely Filing</u>

A Protest may be filed in an application in accordance with 37 C.F.R. § 1.291(b) provided it is "filed prior to the date the application was published under [37 C.F.R.] § 1.211...." Based upon information and research, 37 C.F.R. § 1.211 is the rule that governs publication of pending patent applications by the U.S. Patent Office. As such, it is understood that 37 C.F.R. § 1.291(b) allows a Protest to be filed during the pendency of a U.S. application as long as it is filed prior to the time the application is published by the U.S. Patent Office.

Although the '590 application was published by the International Bureau on October 14, 2004 (*see* the '477 publication (Exhibit A)), the Patent Office has interpreted the term "published" (as found in 37 C.F.R. § 1.211) at 65 Fed. Reg. 57039, stating that "[a]n English language international application designating the United States and published under PCT Article 21(2) is <u>not</u> an application for patent which has been published under 35 U.S.C. § 122(b) (emphasis added)." As such, it is submitted that the '477 publication by the International Bureau is not a publication that would prohibit the entry of this paper in the pending protested application.

B. Identifying Information for Protested Application

As requested by M.P.E.P. § 1901.03, the following statements are believed to be accurate and relevant to the pending protested application:

- (A) Applicants: William DOWNS and George A. FARTHING, Jr.;
- (B) <u>Application No.</u>: 10/591,855 (U.S. National Stage of International Application No. PCT/US2005/009590);
 - (C) International Filing Date: March 21, 2005;

(D) <u>Title</u>: BROMINE ADDITION FOR THE IMPROVED REMOVAL OF MERCURY FROM FLUE GAS;

(E) Group Art Unit: Unknown;

(F) Examiner: Unknown; and

(G) Status: Papers filed on September 5, 2006, to enter the U.S. National Stage.

C. <u>Duplicate Filing in Lieu of Service</u>

As published, the '590 application identifies the applicant as "The Babcock and Wilcox Company" for all designated states except the U.S. See paragraph (71) on the title page of the '477 publication. Further, the U.S. National Stage application, No. 10/591,855, is not yet available to the public. Accordingly, following reasonable research and inquiry, since the undersigned is unable to ascertain with certainty who the applicant of the pending protested application is for purposes of prosecution in the United States, a duplicate copy of this paper is enclosed herewith for the Office to serve on the applicant in accordance with 37 C.F.R. § 1.248 or on its attorney of record or assignee. The Office is respectfully requested, in accordance with and satisfaction of 37 C.F.R. § 1.291(b), to provide a copy of this paper to the applicant or its attorney of record or assignee as identified in the not-yet-public pending protested application.

D. <u>Notification of First Protest By Real Party in Interest</u>

In accordance with 37 C.F.R. § 1.291(b)(2), the Office is hereby informed that, to the best of the undersigned's knowledge based upon reasonable diligence and inquiry, the present Protest is the first and only Protest filed in the pending protested application on behalf of the Protestor (real party in interest).

E. Scope of Protest

As detailed below, and in satisfaction of 37 C.F.R. §§ 1.291(c)(1), (2), and (3), this paper protests the issuance of any claim that claims priority to or is based on the '590 application, to the extent it is directed to the same or substantially the same subject matter as any claim of the internationally published '590 application, as shown in the '477 publication, on the grounds that all the claims of the '477 publication are believed to be unpatentable over the prior art references provided herewith and listed on the Form PTO/SB/08 filed concurrently herewith. A concise explanation of the relevance of each listed reference to the claims of the published '590 application is set forth below.

II. PATENTABILITY STANDARDS: ANTICIPATION AND OBVIOUSNESS

An application for a patent should not be allowed to issue if it is not patentable over the prior art. *See Hoover Group, Inc. v. Custom Metalcraft, Inc.*, 66 F.3d 299, 302 (Fed. Cir. 1995). In order to anticipate a claim under 35 U.S.C. § 102, a reference must contain all elements of the claim, arranged as in the claim. *See* M.P.E.P. § 2131; *Hybritech v. Monoclonal Antibodies, Inc.*, 802 F.2d 1367, 1379 (Fed. Cir. 1986). The law requires identity between the claimed invention and the prior art disclosure. *See Kalman v. Kimberly-Clark Corp.*, 713 F.2d 760, 771, (Fed. Cir. 1983). Importantly, inherent disclosure in the prior art of the claimed subject matter anticipates the claim just as express disclosure does. *See, e.g., Schering Corp. v. Geneva Pharm., Inc.*, 339 F.3d 1373, 1379 (Fed. Cir. 2003).

Furthermore, in order to establish a *prima facie* case of obviousness under 35 U.S.C. § 103, three basic criteria must be demonstrated. First, there must be some suggestion or motivation, either in the references themselves or in the knowledge generally available to one of ordinary skill in the art, to modify the reference or to combine reference teachings. M.P.E.P. §

2143. Second, there must be a reasonable expectation of success. *Id.* Finally, the prior art reference (or references when combined) must teach or suggest all the claim limitations. *Id.*

III. TECHNICAL BACKGROUND

Coal-fired furnaces are a known source of hazardous air pollutants. A range of processes exist for removing or reducing these pollutants, including mercury emissions, from flue gas to insubstantial or non-hazardous levels.

Mercury can appear in combustion flue gas in two different forms, elemental mercury or oxidized mercury. Elemental mercury is gaseous, insoluble in water, and difficult to capture in pollution control devices downstream from the combustion furnace. Oxidized mercury, however, is soluble in water, adsorbs onto carbon sorbents, and is more easily removed from flue gases.

It is well known in the art that halogens readily oxidize mercury and thus can be useful for removing mercury from flue gases. *See* '590 Application at p. 3 (citing U.S. Patent No. 5,435,980, issued to Felsvang et al. (hereinafter referred to as "Felsvang"); *see also* U.S. Patent No. 1,984,164, issued to Stock (hereinafter referred to as "Stock") (teaching the use of halogens for the purification of air containing mercury)). This disclosed functionality is not limited to any particular halogen, although various references in the art do indicate preferences for certain halogens. *See*, *e.g.*, Felsvang; Stock; and U.S. Patent No. 6,878,358, issued to Vosteen et al. (hereinafter referred to as "Vosteen").

The internationally published '590 application¹ is directed to, and claims the use of bromine-containing compounds to enhance the removal of mercury from coal combustion flue

¹ For purposes of this Protest, the internationally published claims of the '590 application, i.e., those contained in the '477 publication, have been assumed to be identical or substantially (continued...)

gases. According to the '590 application, bromine, added directly to the coal to be combusted or to the combustor furnace, enhances the oxidation of elemental mercury, and thereby enhances the overall removal of mercury in pollution control devices at points downstream of the furnace. '590 application, page 3-4.

IV. ANALYSIS OF THE PUBLISHED CLAIMS OF THE '590 APPLICATION

Further to the above general discussion, certain teachings in the Vosteen patent render each element of each claim of the published '590 application, as found in the '477 publication, either anticipated under 35 U.S.C. § 102 and/or *prima facie* obvious under 35 U.S.C. § 103.

A. <u>Vosteen is Prior Art</u>

The teachings of Vosteen are believed to be prior art under at least 35 U.S.C. § 102(e) to any application based on or claiming priority to the '590 application. Vosteen claims priority to its parent, Patent Application Number 10/202,571, filed on July 24, 2002. As such, in accordance with 37 CFR § 102(e), this filing date is believed to be the effective prior art date for the relevant teachings of Vosteen. *See* Vosteen, title page. The '590 application was internationally filed on March 21, 2005, and claims priority to U.S. Provisional Application No. 60/555,353, filed March 22, 2004. *See* '590 application, title page. As such, the earliest possible effective filing date of the currently pending protested U.S. application is March 22, 2004. Thus, Vosteen has a prior art date in advance of the effective U.S. filing date of the currently pending U.S. application, and constitutes valid prior art.

B. Anticipation of Independent Claim 1 et. seq. of the Published '590 Application

^{(...}continued)

similar to those filed and currently pending in the U.S. Patent Office in the pending protested national stage application.

Claim 1, the only independent claim of the published '590 application, is generally directed to a method of removing mercury from a flue gas during combustion of a fossil fuel. Specifically, claim 1 reads:

A method of removing a portion of the elemental mercury in a flue gas created during the combustion of a fossil fuel, comprising:

providing a bromine containing reagent to said flue gas; promoting the oxidation of elemental mercury with the bromine containing reagent;

creating an oxidized form of mercury from the elemental mercury; and

removing the oxidized mercury from the flue gas.

Vosteen discloses each element of this claimed method. Specifically, Vosteen teaches a process for removing mercury from flue gases of high-temperature plants, including, but not limited to, power stations, in which a bromine compound is added "to the furnace and/or to flue gas in a plant section downstream of the furnace, the temperature during the first contact of the bromine compound with the flue gas being at least 500° C . . . " Vosteen at col. 1, 1. 65 - col. 2, 1. 5. The addition of bromine causes oxidation of the mercury and therefore allows substantial removal of the mercury from flue gases. *Id.* at col. 2, 1. 25-31. Several flue gas cleanup processes, such as wet scrubbing or dry cleanup, are known in the art for removing the oxidized mercury from flue gas. *Id.* at col. 5, 1. 3-7.

Dependent claim 2 of the published '590 application further defines the method of claim 1 by requiring that the fossil fuel combusted be coal. Vosteen expressly recites that the invention disclosed therein can be used in bituminous coal-fired power stations. Vosteen at col. 2, l. 32-37. Furthermore, Vosteen also discloses that the bromine compound can be added directly to the coal to be combusted, upstream of the furnace. *Id.* at col. 4, l. 4-8. That disclosure anticipates claim 3 of the published '590 application.

As discussed above, Vosteen is directed to the process of adding bromine directly to a flue gas, which anticipates dependent claim 4 of the published '590 application. *Id.* at col. 3, 1. 65 - col. 4, 1. 4.

Dependent claims 5-7 of the published '590 application are directed to the method of claim 1, wherein the bromine containing reagent is provided in aqueous, solid, and gaseous form, respectively. Vosteen teaches that "[i]t is not critical for the inventive process what form the bromine supplied is present." *Id.* at col. 2, 1. 54-55.

Additionally, Vosteen teaches that the addition of bromine can be made to a coal mill (believed to also be known in the art as a pulverizer), and therefore anticipates dependent claim 8 of the published '590 application, which further defines the method of claim 1 by adding the step of pulverizing the fossil fuel. Vosteen at col. 4, 1. 7-12.

Claim 12 of the published '590 application is directed to the method of claim 1, wherein a substantial portion of the elemental mercury is oxidized. Vosteen teaches that an object of the invention is "to provide a process for . . . the substantially complete removal of mercury . . . from flue gases" Vosteen at col. 1, 1. 50-54. According to Vosteen, this can be accomplished by the "substantial, essentially complete oxidation of the mercury" *Id.* at col. 2, 1. 29-31.

Claims 13-16 of the published '590 application all relate to systems and apparatus according to claim 1 for the removal of oxidized mercury from flue gas. These claims are fully anticipated by the disclosure of Vosteen at col. 4, l. 66 - col. 5, l. 39, including a wet flue gas scrubbing system (anticipating claim 13), a dry emission control system (anticipating claim 14), and a fixed-bed adsorber based on activated carbon (anticipating claims 15-16).

C. Obviousness of the Dependent Claims of the Published '590 Application

Claim 10 of the published '590 application is dependent on claim 2, and further defines the method wherein the coal is treated with up to about 1000 ppm of bromine. Claim 11 of the published '590 application, dependent on claim 10, contains the further limitation that the coal is treated with between about 100 and about 200 ppm of bromine. These claims are believed to be obvious to the skilled artisan in view of the teachings of Felsvang in combination with those of Vosteen and Stock. Felsvang is directed to the removal of mercury from flue gas, and teaches the addition of a chloride or chloride containing material to coal before or during combustion, such that the total amount of chloride in the flue gas is 150 ppm. Felsvang at col. 4, 1. 39-62. Although Felsvang does not disclose treating the coal with bromine, substitution of bromine for chlorine would have been obvious to a person of ordinary skill in the art due to the interchangeability of halogens for oxidizing mercury, as disclosed in Stock. *See* Stock at col. 1, 1. 33-38.

As a result, it would have been obvious to combine the relevant teachings in Felsvang with those in Stock and Vosteen to add bromine to the coal in the amounts disclosed in claims 10 and 11 of the '590 application. *See* Felsvang at col. 4, 1. 39-62; Stock at col. 1, 1. 33-38; and Vosteen at col. 4, 1. 4-12 and 34-48.

Finally, claim 9 of the published '590 application merely recites a common practice in the relevant art that would have been obvious to a person of ordinary skill in the art practicing the teachings of Vosteen, i.e., to pulverize the fossil fuel after treating the fossil fuel. Coal is routinely crushed or pulverized before entering a coal-fired furnace, and often the pulverizer is on top of the furnace, several hundred feet above ground. Therefore, it would have been obvious

to a person of ordinary skill in the art who wanted to add a chemical to the coal to add it upstream of the pulverizer.

V. CONCLUSION

For at least the foregoing reasons, it is respectfully submitted that each of the claims of the published '590 application, as shown in the '477 publication, and any claim based on or claiming priority to the '590 application that is directed to the same or substantially the same subject matter as the claims of the published '590 application, are unpatentable.

If there is a fee due in connection with this Protest, please charge the fee to Deposit Account No. 06-0916.

Respectfully submitted,

FINNEGAN, HENDERSON, FARABOW, GARRETT & DUNNER, L.L.P.

Dated: December 20, 2006

Michele C. Boscl Reg. No. 40,524

(202) 408-4193

Exhibit A

(19) World Intellectual Property Organization International Bureau



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22 March 2004 (22.03.2004) US

(71) Applicant (for all designated States except US): THE BABCOCK & WILCOX COMPANY [US/US]; 20 S. Van Buren Avenue, Barberton, OH 44203-0351 (US).

(72) Inventors; and

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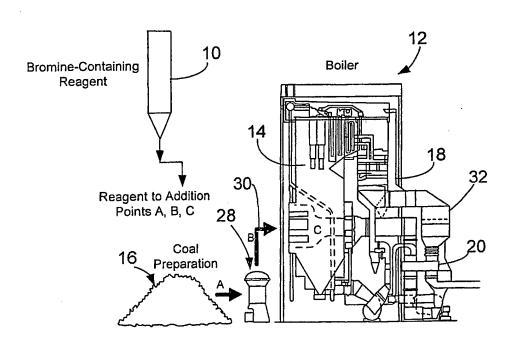
- (74) Agent: MARICH, Eric; The Babcock & Wilcox Company, Patent Department, 20 S. Van Buren Avenue, Barberton, OH 44203-0351 (US).
- (81) Designated States (unless otherwise indicated, for every kind of national protection available): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BW, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NA, NI, NO, NZ, OM, PG, PH, PL, PT, RO, RU, SC, SD, SE, SG, SK, SL, SM, SY, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, YU, ZA, ZM, ZW.
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Published:

with international search report

[Continued on next page]

(54) Title: BROMINE ADDITION FOR THE IMPROVED REMOVAL OF MERCURY FROM FLUE GAS



(57) Abstract: Bromine-containing compounds (10), added to the coal (16), or to the boiler combustion furnace (14), are used to enhance the oxidation of mercury, thereby enhancing the overall removal of mercury in downstream pollution control devices. The method is applicable to utility power plants equipped with wet FGD systems, as well as those plants equipped with spray dryer absorber FGD systems.

[0001] BROMINE ADDITION FOR THE IMPROVED REMOVAL
[0002] OF MERCURY FROM FLUE GAS

[0003] FIELD AND BACKGROUND OF THE INVENTION

[0004] Emissions Standards, as articulated in The Clean Air Act Amendments of 1990 as established by the U.S. Environmental Protection Agency (EPA), required assessment of hazardous air pollutants from utility power plants. In December 2000 the EPA announced their intention to regulate mercury emissions from coal-fired utility boilers. Coal-fired utility boilers are a known major source of anthropogenic mercury emissions in the United States. Elemental mercury and many of its compounds are volatile and will therefore leave the boiler as trace constituents in boiler flue gases. Some of these mercury constituents are insoluble in water, which renders them difficult to capture in conventional wet and dry scrubbers. Thus new methods and processes are needed to capture these trace constituents from boiler flue gases.

[0005] Mercury appears in coal combustion flue gases in both solid and gas phases (particulate-bound mercury and vapor-phase mercury, respectively). The so called particulate-bound mercury is really vapor-phase mercury adsorbed onto the surface of ash or carbon particles. Due to the high volatility of mercury and many of its compounds, most of the mercury found in flue gases is vapor-phase mercury. Vapor-phase mercury can appear as elemental mercury (elemental, metallic mercury vapor) or as oxidized mercury (vapor-phase species of various compounds of mercury). Speciation, which refers to the form of mercury present, is a key parameter in the development and design of mercury control strategies. All efforts to devise new control strategies for mercury emissions from power plants must focus on this characteristic of mercury.

Particulate collectors in use at electric utility plants, most commonly [0000] electrostatic precipitators (ESP) or fabric filters (FF), sometimes called baghouses, provide high-efficiency removal of particulate-bound mercury. Fabric filters tend to exhibit better particulate laden mercury removal than ESPs by providing a filter cake upon which to trap the particulate mercury as the flue gas passes through said filter cake. If the filter cake also contains constituents that will react with mercury such as unreacted carbon or even activated carbon, then the filter cake can act as a site to facilitate gas-solid reactions between the gaseous mercury and the solid carbon particles. If a power plant is equipped with a Flue Gas Desulfurization System (FGD) then either wet scrubbers or spray dryer absorbers (SDA) can remove significant amounts of oxidized mercury. Oxidized mercury, typically appearing in the form of mercuric chloride, is soluble in water, making it amenable to removal in sulfur dioxide scrubbers. Elemental mercury, insoluble in water, is less likely to be scrubbed in conventional scrubbers. Removal of elemental mercury, therefore, remains an important issue in the search for cost-effective mercury control techniques.

Numerous studies have been, and continue to be, conducted to [0007] develop cost-effective approaches to the control of elemental mercury. Many of the studies have focused on the injection of a carbonaceous sorbent (e.g., powdered activated carbon, or PAC) into the flue gas upstream of the dust collector to adsorb vapor-phase mercury. The sorbent, and its burden of adsorbed mercury, are subsequently removed from the flue gases in a downstream particulate collector. Adsorption is a technique that has often been successfully applied for the separation and removal of trace quantities of undesirable components. PAC injection is used, commercially, to remove mercury from municipal waste combustor exhaust gases. PAC injection removes both oxidized and elemental mercury species, although removal efficiencies are higher for the oxidized form. Although this approach appeared attractive in early work, the economics of high injection rates can be prohibitive when applied to coal-fired utility plants. More refined studies are now in progress to define more precisely what can and cannot be achieved with PAC. Still other studies seek to enhance PAC technology. One technique subjects the PAC to an impregnation process wherein elements such as iodine or sulfur are incorporated into the carbonaceous sorbent. Such processes can yield sorbents that more strongly bond with adsorbed mercury species, but also result in significantly higher sorbent cost.

The speciation of vapor-phase mercury depends on coal type. Eastern [0008] U.S. bituminous coals tend to produce a higher percentage of oxidized mercury than do western subbituminous and lignite coals. Western coals have low chloride content compared to typical eastern bituminous coals. It has been recognized for several years that a loose empirical relationship holds between the chloride content of coal and the extent to which mercury appears in the oxidized form. Fig. 1 (Source: Senior, C.L. Behavior of Mercury in Air Pollution Control Devices on Coal-Fired Utility Boilers, 2001) illustrates the relationship between coal chlorine content and vapor-phase mercury speciation. An important reason for the significant scatter in the data of Fig. 1 is that mercury oxidation depends in part on the specific characteristics of the boiler as well as the fuel. The mercury oxidation reactions proceed by both homogeneous and heterogeneous reaction mechanisms. Factors such as boiler convection pass and combustion air preheater temperature profiles, flue gas composition, fly ash characteristics and composition, and the presence of unburned carbon have all been shown to affect the conversion of elemental mercury to oxidized mercury species.

[0009] Felsvang et al. (U.S. Patent No. 5,435,980) teaches that the mercury removal of a coal-fired system employing an SDA system can be enhanced by increasing the chlorine-containing species (e.g., hydrogen chloride) in the flue gases. Felsvang et al. further teaches that this can be accomplished through the addition of a chlorine-containing agent to the combustion zone of the boiler, or through the injection of hydrochloric acid (HCI) vapor into the flue gases upstream of the SDA. These techniques are claimed to improve the mercury removal performance of PAC when used in conjunction with an SDA system.

[0010] SUMMARY OF THE INVENTION

[0011] It is an object of this invention to invention to yield significant technical and commercial advantages over the prior art. The present inventors have determined through experimental testing that the use of bromine-containing compounds, added to the coal, or to the boiler combustion furnace, are significantly

more effective than chlorine-containing compounds in enhancing the oxidation of mercury, thereby enhancing the overall removal of mercury in downstream pollution control devices. Second, the technique is applicable to utility power plants equipped with wet FGD systems, as well as those plants equipped with SDA systems. Wet FGD is the sulfur dioxide removal system of choice for most coal-fired utilities around the world. Approximately 25% of the coal-fired electric power plants in the U.S. are equipped with wet FGD systems.

[0012] The various features of novelty which characterize the invention are pointed out with particularity in the claims annexed to and forming a part of this disclosure. For a better understanding of the present invention, its operating advantages and the specific benefits attained by its uses, reference is made to the accompanying drawings and descriptive matter in which preferred embodiments of the invention are illustrated.

[0013] BRIEF DESCRIPTION OF THE DRAWINGS

- [0014] Fig. 1 is a graph illustrating the relationship between coal mercury content and mercury speciation for U.S. coals;
- [0015] Fig. 2 is a schematic illustration of a first embodiment of the present invention involving bromine addition for the improved removal of mercury from flue gases;
- [0016] Fig. 3 is a graph of test data illustrating the effect of the addition of a particular halogen, calcium bromide, CaBr₂, on the total vapor-phase mercury produced during the combustion of coal, according to the present invention;
- [0017] Fig. 4 is a schematic illustration of a coal-fired electric utility plant configuration comprising a boiler equipped with an SDA and a downstream particulate collection means such as a fabric filter (FF) or an electrostatic precipitator (ESP);
- [0018] Fig. 5 is a schematic illustration of a coal-fired electric utility plant configuration comprising a boiler equipped with a downstream particulate collection means such as a fabric filter (FF) or an electrostatic precipitator (ESP); and

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Fig. 6 is a schematic illustration of a coal-fired electric utility plant [0019] configuration comprising a boiler equipped with a downstream particulate collection means such as a fabric filter (FF) or an electrostatic precipitator (ESP) and a wet flue gas desulfurization (FGD) system.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS [0020]

Referring to the drawings generally, wherein like numerals designate [0021] the same or functionally similar elements throughout the several drawings, a first embodiment of the present invention is illustrated in Fig. 2. A bromine-containing reagent 10 is added to the boiler 12 combustion furnace 14, either directly or by premixing with the incoming coal 16. Bromine species released during the combustion process enhance the oxidation of mercury as the combustion gases pass through the furnace 14 and, in particular, through the cooler sections of the boiler convection pass 18 and combustion air preheater 20. The increased fraction of mercury appearing in the oxidized form enhances mercury removal in downstream: pollution control systems such as wet 22 and SDA 24 FGD systems, and PAC injection systems. As is described herein, experimental results indicate that bromine addition also results in an increased fraction of particulate-bound mercury. This enhances removal of mercury across particulate collectors 26 such as fabric filters (FF) and electrostatic precipitators (ESP).

The removal of elemental mercury from coal combustion gases [0022] generated by electric utility plants through the application of a conventional PAC injection process is very expensive. The present invention promises to significantly reduce the cost of mercury removal at coal-fired electric plants in two ways. First, increasing the fraction of mercury appearing in the oxidized and particulate-bound forms enhances the removal of mercury in conventional pollution control systems. such as particulate collectors 26 and wet 22 and SDA 24 FGD systems. This reduces, or may eliminate entirely, the need for PAC injection to remove elemental mercury. Second, the increased fraction of oxidized mercury also enhances the removal of mercury across a PAC injection process, due to the higher reactivity of oxidized mercury with PAC.

The present invention was tested in a 5 million Btu/hr Small Boiler [0023]T Simulator (SBS) Facility. The SBS was fired at approximately 4.3 million Btu/hr with a western U.S. subbituminous coal. During the tests, flue gases exiting the SBS boiler first passed through a spray dryer absorber (SDA) for removal of sulfur dioxide, and then through a fabric filter (FF) for removal of fly ash and spent sorbent from the SDA FGD system.

An aqueous solution of calcium bromide (CaBr₂) was injected into the [0024]combustion chamber 14 through a coal burner (not shown). Fig. 3 illustrates the removal of mercury across the SDA/FF system. It can be seen that upon injection of the calcium bromide, the vapor-phase mercury exiting the system dropped from its initial value of approximately 6 µg/dscm to about 2 µg/dscm. It can also be seen that the vapor-phase mercury at the system inlet also drops upon addition of the calcium This is due to the fact that the calcium bromide also enhances the formation of particulate-bound mercury (particulate-bound mercury does not appear on the chart, since the on-line mercury analyzer being used only detects vaporphase mercury species.). These results identify that the current invention can offer a cost-effective method of removing elemental mercury from coal combustion flue gases.

[0025] In the preferred embodiment, an aqueous solution of calcium bromide is sprayed onto the crushed coal 16 before the coal 16 is pulverized for combustion. The aqueous solution is easily handled and metered onto the coal 16, coal pulverizers 28 intimately mix the bromide reagent 10 with the coal 16, and the pulverized coal conveying system 30 to the several coal burners (not shown) ensures an even distribution of the reagent 10 across the boiler furnace 14. There are many alternative ways to implement the invention as would be apparent to one of skill in the art. Based upon the tests performed, it is believed that adequate mercury removal can be achieved when the coal 16 is treated with up to about 1000 ppm of bromine from the bromine containing reagent 10; particularly between about 100 and about 200 ppm of bromine from the bromine containing reagent 10. As will be appreciated by those skilled in the art, some non-zero amount of bromine must be supplied in order to apply the principles of the invention; the upper limit of the range

is, as a practical matter, limited by the possible increased corrosion potential which might be created.

[0026] In another embodiment the coal-fired boiler fuel 16 may include bituminous, subbituminous, and lignite coals and blends, thereof.

[0027] In yet another embodiment, the bromine-containing reagent 10 could comprise, but is not limited to, alkali metal and alkaline earth metal bromides, hydrogen bromide (HBr) or bromine (Br₂).

[0028] In yet another embodiment the bromine-containing reagent 10 may be fed to the boiler combustion zone 14 in gaseous, liquid, or solid form.

[0029] In yet another embodiment, the electric utility plant configurations may include plants equipped with an SDA 24 and particulate collector 26 (FF or ESP) (Fig. 4), a particulate collector 26 (FF or ESP) (Fig. 5), or a wet 22 FGD and particulate collector 26 (FF or ESP) (Fig. 6).

[0030] In yet another embodiment, the invention may be utilized in a coal-fired plant equipped with a selective catalytic reduction (SCR) system 32 for the control of nitrogen oxides, as SCR catalysts have been shown to promote the oxidation of elemental mercury if the correct species (in this case bromine species) are present in the flue gases.

[0031] In yet another embodiment, mercury removal may be further enhanced by utilizing a sorbent injection system in conjunction with the present invention. Such carbonaceous sorbents include, but are not limited to, powdered activated carbon (PAC), carbons and chars produced from coal and other organic materials, and unburned carbon produced by the combustion process itself.

[0032] While specific embodiments of the invention have been shown and described in detail to illustrate the application of the principles of the invention, those skilled in the art will appreciate that changes may be made in the form of the invention covered by the following claims without departing from such principles. For example, the present invention may be applied to new fossil-fueled boiler construction which requires removal of mercury from flue gases produced thereby, or to the replacement, repair or modification of existing fossil-fueled boiler installations. In some embodiments of the invention, certain features of the invention may sometimes be used to advantage without a corresponding use of the other features.

Accordingly, there are other alternative embodiments which would be apparent to those skilled in the art and based on the teachings of the present invention, and which are intended to be included within the scope and equivalents of the following claims of this invention.

[0033] CLAIMS

[0034] We claim:

[0035] 1. A method of removing a portion of the elemental mercury in a flue gas created during the combustion of a fossil fuel, comprising:

providing a bromine containing reagent to said flue gas;

promoting the oxidation of elemental mercury with the bromine containing reagent;

creating an oxidized form of mercury from the elemental mercury; and removing the oxidized mercury from the flue gas.

- [0036] 2. The method according to claim 1, wherein the fossil fuel is coal.
- [0037] 3. The method according to claim 1, wherein the step of providing the bromine containing reagent comprises the step of treating the fossil fuel with the bromine containing reagent prior to combustion.
- [0038] 4. The method according to claim 1, comprising the step of treating the flue gas with the bromine containing reagent.
- [0039] 5. The method according to claim 1, wherein the bromine containing reagent is provided in an aqueous form.
- [0040] 6. The method according to claim 1, wherein the bromine containing reagent is provided in a solid form.
- [0041] 7. The method according to claim 1, wherein the bromine containing reagent is provided in a gaseous form.
- [0042] 8. The method according to claim 3, further comprising the step of pulverizing the fossil fuel.

- [0043] 9. The method according to claim 8, wherein the pulverizing step occurs after the treating step.
- [0044] 10. The method according to claim 2, wherein the coal is treated with up to about 1000 ppm of bromine from the bromine containing reagent.
- [0045] 11. The method according to claim 10, wherein the coal is treated with between about 100 and about 200 ppm of bromine from the bromine containing reagent.
- [0046] 12. The method according to claim 1, wherein a substantial portion of the elemental mercury in the flue gas is oxidized.
- [0047] 13. The method according to claim 1, further comprising the step of using a wet flue gas desulfurization apparatus to remove a substantial portion of the oxidized mercury from the flue gas.
- [0048] 14. The method according to claim 1, further comprising the step of using a spray dryer flue gas desulfurization apparatus to remove a substantial portion of the oxidized mercury from the flue gas.
- [0049] 15. The method according to claim 1, further comprising the step of using a sorbent injection system to remove a substantial portion of the oxidized mercury from the flue gas.
- [0050] 16. The method according to claim 15, wherein the sorbent comprises powdered activated carbon.

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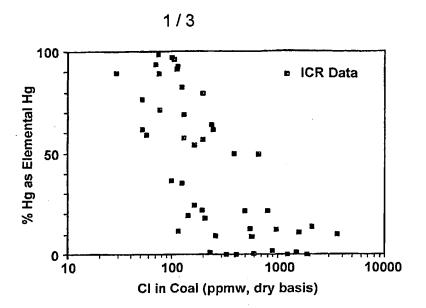


FIG. 1

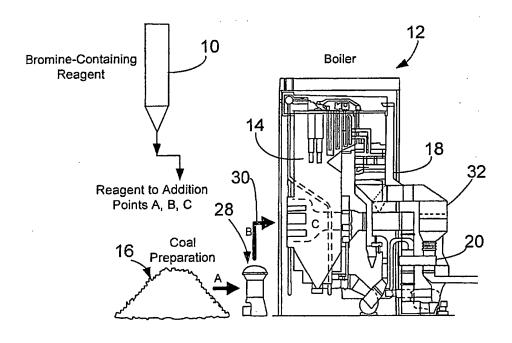


FIG. 2

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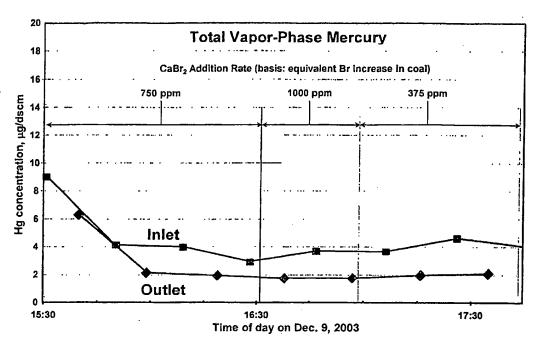


FIG. 3

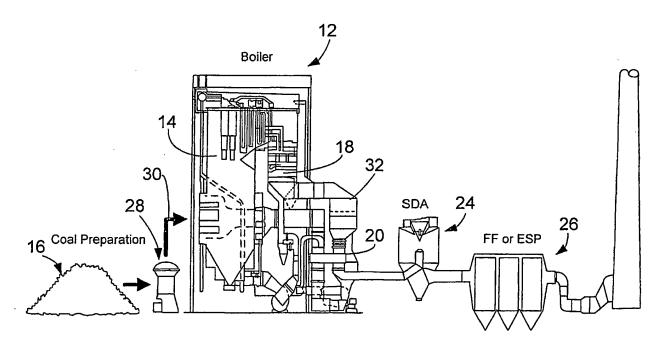
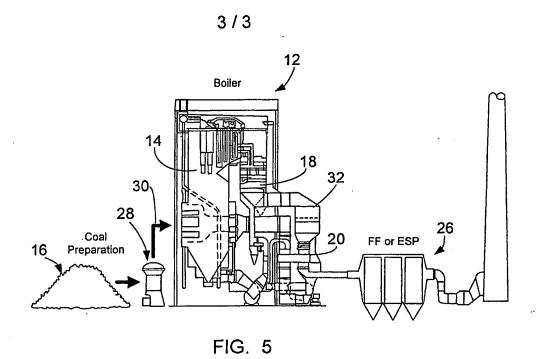


FIG. 4



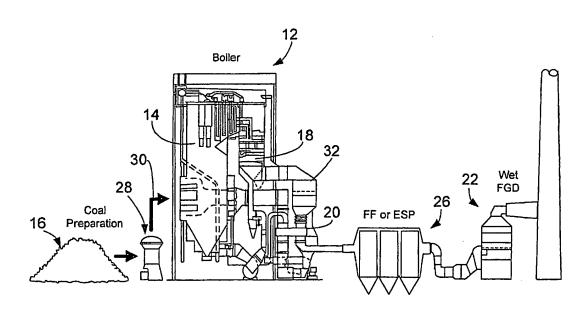


FIG. 6

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US05/09590

A. CLASSIFICATION OF SUBJECT MATTER IPC(7) : B01D 47/00, 53/54: B01J 8/00; A62D 3/00 US CL : 423/210,212,213,99,101; 588/404,407,412				
	International Patent Classification (IPC) or to both nat	ional classification and IPC		
B. FIELI	OS SEARCHED			
Minimum documentation searched (classification system followed by classification symbols) U.S.: 423/210,212,213,99,101; 588/404,407,412				
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched				
Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)				
C. DOCUMENTS CONSIDERED TO BE RELEVANT				
Category *	Citation of document, with indication, where appropriate, of the relevant passages		Relevant to claim No.	
A	US 5,435,980 A (FELSVANG et al.) 25 July 1995 (25.07.1995), entire document. 1-16		1-16	
. A	US 4,196,173 (DEJONG et al.) 1 April 1980 (01.04.1980), entire document.		1-16	
Α	US 3,849,267 (HILGEN et al.) 19 November 1974 (19.11.1974), entire document.		1-16	
Α	US 4,889,698 (MOLLER et al.) 26 December 1989 (26.12.1989), entire document.		1-16	
Α	US 5,695,726 (LERNER, Bernard J.) 9 December 1997 (09.12.1997), entire document.		1-16	
Α	US 6,375,909 B1 (DANGTRAN et al.) 23 April 2002 (23.04.2002), entire document. 1-16		1-16	
Α	US 6,136,281 A (MEISCHEN et al.) 24 October 2000 (24.10.2000), entire document.		1-16	
Α	US 6,284,208 B1 (THOMASSEN, Thomas) 04 September 2001 (04.09.2001), entire document.		1-16	
Further	documents are listed in the continuation of Box C.	See patent family annex.		
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Date of the actual completion of the international search Date of mailing of the international search report		ch report		
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